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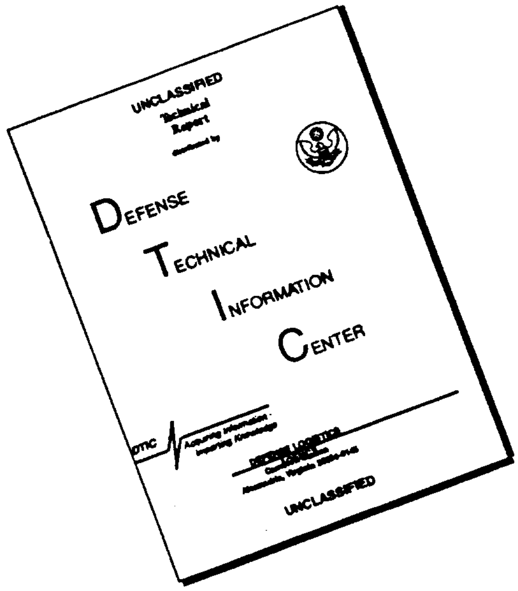

Photothermal Investigations of GaAs-Based Materials

and Quantum-Well Structures

Final Report on AFOSR Grant No. F49620-92-J-0381

Executive Summary

Photothermal deflection spectroscopy has been applied to GaAs samples with the ultimate goal of measuring the optoelectronic and thermal transport properties of GaAs based multiple quantum wells by this technique. Theoretical models have been developed as well as experimental investigations have been carried out. Both pulsed as well as cw (modulated) excitations have been considered.

For the pulsed case, the theoretical model predicts a photothermal deflection signal which is a complicated function of the thermal diffusivities, thermal conductivities, and the thicknesses of the sample, backing (on which the sample is mounted), and the fluid (in which the probe beam propagates). By an analysis of the deflection signal both the thermal diffusivity and the optical absorption coefficient of the sample can be determined. The experiment has verified the theory in general terms, but quantitative agreement has not yet been achieved. These results will be described fully in the Ph.D. dissertation of George Bennis (University of Arkansas, 1997).

For the cw case, the theory predicts that the optical absorption coefficient and the thermal transport properties can be obtained either from the magnitude or the phase of the signal. Moreover, the cw technique offers a way of measuring the thermal transport properties which is much more reliable than the pulsed technique. The experimental signal shapes agree with theoretical predictions in general, however, a quantitative agreement has not yet been achieved. Some of these results are described in William H. Leach's M.S. thesis (University of Arkansas, 1996) and more will be described in his Ph.D. dissertation (University of Arkansas, 1998).

Photothermal Investigations of GaAs-Based Materials and Quantum-Well Structures

Final Report on AFOSR Grant No. F49620-92-J-0381

We have applied photothermal spectroscopy to GaAs materials with the ultimate goal of measuring the thermal transport and optoelectronic properties of multiple quantum well samples.

Figure 1 shows the basic idea behind a photothermal experiment. The sample of thickness ℓ is assumed to be deposited on a backing material of thickness ℓ_b, and is in contact with a fluid of thickness ℓ_f. This model also works for a free-standing sample, in which case the backing is replaced by the fluid. A laser beam is incident on the sample from the right. The wavelength of the laser beam is tuned to a region where the sample has a significant optical absorption. The backing and the fluid are assumed to be transparent at this wavelength. The optical energy quickly appears as heating of the sample. The heat diffuses laterally in the sample, as well as backward and eventually into the backing. The heat also diffuses into the fluid where a refractive index gradient is produced. The refractive index gradient is detected by a second, weaker laser beam, called the probe beam. The fluid is transparent to the probe beam also. The probe beam gets deflected due to the nonuniform refractive index created by the heating of the fluid, much like bending of light rays in mirage, and can be detected by a position-sensitive detector. The deflection has two components, Θ_n, normal to the sample surface, and Θ_t, tangential to the sample surface. We generally measure Θ_n because it is stronger. The size of the photothermal deflection signal, Θ_n, is proportional to the amount of heat produced, and thus it is a sensitive measure of the optical absorption coefficient of the sample. Measurement of the optical absorption coefficient as a function of the wavelength of the pump laser and temperature of the sample would provided a wealth of information on the optoelectronic properties of the sample. As one might imagine, the photothermal deflection signal depends, in addition to the optical absorption coefficient, on

the thermal transport properties of the sample. The photothermal technique thus offers a convenient way of measurement of the thermal transport properties. Such measurements are valuable in themselves because of the problem of heat dissipation in modern semiconductor devices.

We have developed a detailed theory of photothermal signal under the conditions of Fig. 1. The signal is given by

$$\Theta_n = \frac{1}{n_0}\frac{\partial n}{\partial T}\int_{-\infty}^{\infty}\frac{\partial T_f}{\partial z}dx \tag{1}$$

where T_f is the temperature of the fluid at the point of measurement, $\partial n/\partial T$ is the gradient of refractive index of the fluid with respect to temperature, and n_0 is the refractive index of the fluid at the ambient temperature.

The temperature distribution can be determined by solving the differential equations relating to the conduction of heat. For the three region model of the sample described above and shown in Fig. 1 the heat diffusion equations in cylindrical coordinates are:

$$\frac{\partial^2\phi_f(r,z,t)}{\partial r^2}+\frac{1}{r}\frac{\partial\phi_f(r,z,t)}{\partial r}+\frac{\partial^2\phi_f(r,z,t)}{\partial z^2}=\frac{1}{D_f}\frac{\partial\phi_f(r,z,t)}{\partial t},\quad 0\le z\le \ell_f \tag{2}$$

$$\frac{\partial^2\phi_s(r,z,t)}{\partial r^2}+\frac{1}{r}\frac{\partial\phi_s(r,z,t)}{\partial r}+\frac{\partial^2\phi_s(r,z,t)}{\partial z^2}=\frac{1}{D_s}\frac{\partial\phi_s(r,z,t)}{\partial t}-A(r,t)e^{\alpha z},\quad -\ell\le z\le 0 \tag{3}$$

$$\frac{\partial^2\phi_b(r,z,t)}{\partial r^2}+\frac{1}{r}\frac{\partial\phi_b(r,z,t)}{\partial r}+\frac{\partial^2\phi_b(r,z,t)}{\partial z^2}=\frac{1}{D_b}\frac{\partial\phi_b(r,z,t)}{\partial t},\quad -\ell-\ell_b\le z\le -\ell \tag{4}$$

ϕ_f,ϕ_s, and ϕ_0 represent the complex expressions for the temperature, whereas T_f, T_s, and T_b represent their real parts, respectively; the subscripts f, s and b refer to the fluid, sample, and backing regions of the model, respectively. The constants D_f, D_s, and D_b are the thermal diffusivities of the appropriate materials while k_f, k_s, and k_b are the thermal conductivities.

The constants ℓ, ℓ_f, and ℓ_b are the dimensions along the z axis of the sample, fluid and backing, respectively, as shown in Fig. 1. The source term appears only in Eq. (3). We have considered both cw and pulsed excitation. The source term $A(r,t)$, assuming that the pump beam is a continuous wave laser, chopped at a frequency $f = \omega/2\pi$, and having a Gaussian profile is defined as,

$$A(r,t) = \frac{\alpha P \eta}{\pi k_s a^2} \exp\left(- \frac{2\mathrm{r}^2}{\mathrm{a}^2} \right) \times (1 + \mathrm{e}^{\mathrm{i}\omega t}) \tag{5}$$

where the peak pump beam power is P, the pump beam radius is a, and the optical absorption coefficient is α. The efficiency of conversion of pump energy into heat is represented by η. The source term for pulsed excitation is given by

$$A(r,t) = \frac{\alpha P \eta}{\pi k_s a^2} \exp\left(- \frac{2\mathrm{r}^2}{\mathrm{a}^2} \right) \times \delta(\mathrm{t}) \tag{6}$$

where $\delta(t)$ is the delta function and P is the peak power in the laser pulse. The laser pulse is assumed to be very short (delta pulse). Equations (2)-(4) are solved subject to the boundary conditions that the temperature as well as the heat flow across each interface be continuous. We have conducted the experiments as well as developed the theoretical model for both cw (modulated) and pulsed excitations. In the following we describe the results of both investigations separately.

I. <u>Pulsed Excitation</u>

The solution of Eqs. (2)-(4) for the source term given by Eq. (6), to the best of our knowledge, is not available in the literature. We have solved these equations, and the solution is rather involved[1]. The solution involves double integration in a complex plane, and the integrations must be performed numerically. The results are subject to large numerical errors unless extreme care is exercised. The resulting temperature depends in a rather complex manner on the thermal transport properties and thicknesses of the backing, sample, and the fluid.[1] Figures 2 and 3 show the effects of varying the thermal diffusion constant and

the optical absorption coefficient of the sample (assumed to be GaAs) on the photothermal deflection signal, respectively. The signals consist of the deflection of the probe beam shortly after the pump laser pulse, and the deflection gradually goes to zero as the heat diffuses out of the probe beam region. Figure 2 shows the effect of the sample diffusivity. The solid line is for the nominal values of D_s, while the dotted and dashed lines are for values of D_s then times smaller and ten times larger, respectively. Figure 3 shows the effect of varying the sample absorption coefficient. The solid line is for the nominal value of the absorption coefficient for photon energies larger then the bandgap, whereas the other curves are for larger or smaller values of the absorption coefficient. These curves show that in principle it is possible to determine D_s and α from the magnitude of the signal, however, the dependences of Θ_n on both α and D_s are highly non-linear and in certain range, it appears difficult to disentangle the variation of Θ_n on D_s from that on α. The wavelength dependence of the signal, however, could still provide the useful information on α.

Figure 4 shows a sketch of the experiment. A Ti-sapphire laser pumped by a Q-switched doubled Nd:YAG laser (wavelength range 780nm - 950nm, $\sim$ 7ns pulse length) is used as the pump laser. An HeNe laser grazing the surface of the sample is used as the probe laser. The sample is a free-standing wafer of GaAs and it is immersed in $CC\ell_4$ (that is, both the fluid and the 'backing' are $CC\ell_4$). The deflection of the probe beam is measured by a bi-cell detector in conjunction with a difference amplifier. In our earlier experiments the signal pulse was captured by a transient digitizer but more recently we have used a digital oscilloscope to capture the signal.

Figure 5 shows a typical signal (solid line). The theoretically predicted curve is plotted on the same graph (dashed line). The theoretical curve has been computed for the exact conditions pertaining to the experiment and has no adjustable parameters. The experimental

signal is approximately a factor of two smaller and it is broader. At this time we are trying to understand this anomaly. Once this anomaly is resolved, we will be able to publish these results.

II. CW Excitation

The theory of photothermal spectroscopy with modulated excitation is available in the literature.[2] The results have a simple physical interpretation. As a result of the optical excitation 'thermal waves' are set up in the sample which reflect at each interface. Thermal waves are also setup in the fluid with a characteristic attenuation constant given by the thermal diffusivity of the fluid and the modulation frequency of the laser.

The results cannot be obtained in a closed form and must be obtained numerically. We have written a computer program to evaluate the expressions and some of the results are shown below.[3] Figures 6 and 7 show the amplitude and phase of the normal deflection signal as a function of the optical absorption coefficient of the sample for several values of the sample diffusivity, respectively. These figures show that, in principle, it is possible to determine optical absorption coefficient α and the sample diffusivity D_s by measurements of either the amplitude on the phase of the deflection signal. However, since both D_s and α affect the magnitude and phase of Θ_n, an unambiguous determination of D_s and α from these measurements is difficult. Therefore we have explored another method, which is independent of the value of α, for the determination of D_s. The deflection signal Θ_n can be resolved into an in-phase and a quadrature component. Figure 8 shows a theoretical plot of the in-phase component of Θ_n as a function of the lateral coordinate y. The distance between the zero-crossing points of these curves depends, not only on the frequency of modulation f as shown, but also on the thermal diffusivity of the sample. A plot of the distance between the crossing points as a function of $f^{-1/2}$ yields a straight line whose slope is proportional to D_s. This determines D_s uniquely, and then α can be determined from the amplitude and/or the phase of the deflection signal.[3]

Figure 9 shows the experimental arrangement for the cw experiment. Pump beam consists of the radiation from a Ar^+ laser pumped ring dye laser operating in the near-IR range. The probe beam is supplied by a HeNe laser. The GaAs sample is placed in a cuvette filled with $CC\ell_4$. The probe beam travels parallel to the sample surface and is detected by a bi-cell or a quadrant detector. The pump beam is chopped by a mechanical chopper. The detector output is measured by a difference amplifier and the signal is detected by a digital lock-in amplifier.

Figure 10 shows a typical result. The rms value of the deflection signal Θ_n is plotted against the y-position of the probe beam, and the shape of the curve agrees with our expectations. Figure 11 shows the magnitude of the signal at its peak plotted against the modulation frequency. The solid curve shows the theoretical prediction (with no adjustable parameters), and the squares show the experimental results. The experimental values have been divided by a factor of two for the ease of plotting theory and experiment on the same graph. We note that the experimental value is higher than the theoretical value by approximately a factor of two at the modulation frequency of 2Hz, and the disagreement between the theory and the experiment gets progressively worse as the frequency is increased. We have not yet been able to determine the cause of this discrepancy – although there are several leads that we are pursuing.[3]

Figure 12 shows the distance between the zero-crossing points of the in-phase portion of Θ_n, y_0, as a function of $f^{-1/2}$. The solid line shows the theoretical prediction, whereas the squares represent the experimental points. Clearly, the slope of the experimental curve is much smaller than predicted. We think that the problem is due to the fact that our sample size (5mm $\times$ 10mm) is comparable to the thermal diffusion length at these low frequencies, whereas the theory assumes it to be of infinite lateral dimensions. Unfortunately,

we are forced to use low modulation frequencies because of the very low value of the thermal diffusion constant for $CC\ell_4$. We will repeat the experiment with a larger sample to test our ideas.

In conclusion, we have done a significant amount of work, both theoretically and experimentally, both with the pulsed excitation and with cw excitation, but have not yet resolved the disagreements between the theory and the experiment. As soon as these disagreements are resolved, we will be able to carry out the proposed measurements of thermal transport and opto-electronic properties.

Reference:

1. Details will be given in Ph.D. dissertaion by George Bennis, University of Arkansas, 1997.

2. D. Fournier and A.C. Boccara in Photothermal Investigations of Solids and Fluids, ed. J.A. Sell, Academic Press, N.Y., 1989.

3. W.H. Leach, M.S. Thesis, University of Arkansas, 1996.

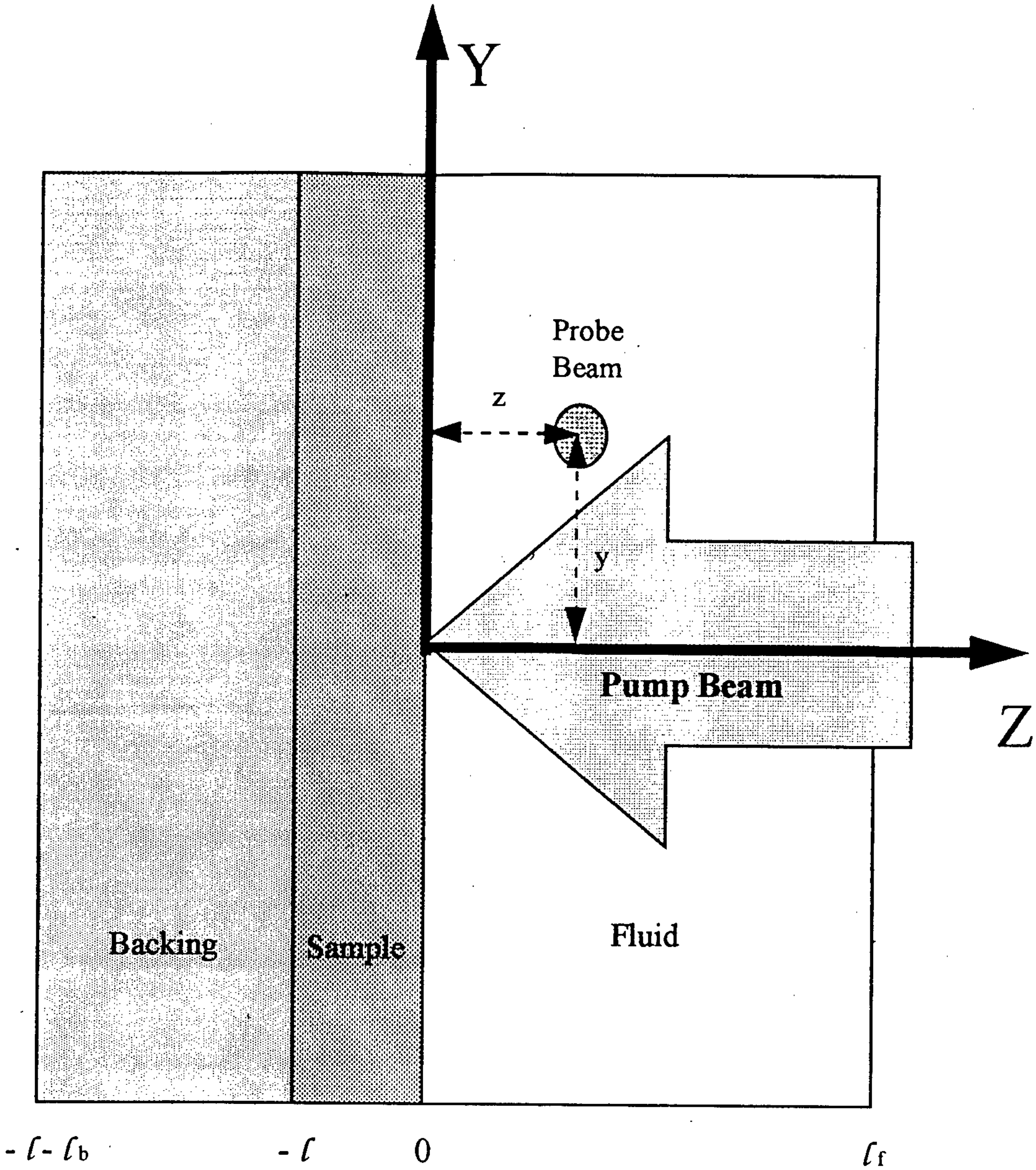

Fig. 1 Diagram of the model for PTDS showing the sample cell, lasers and coordinate axes.

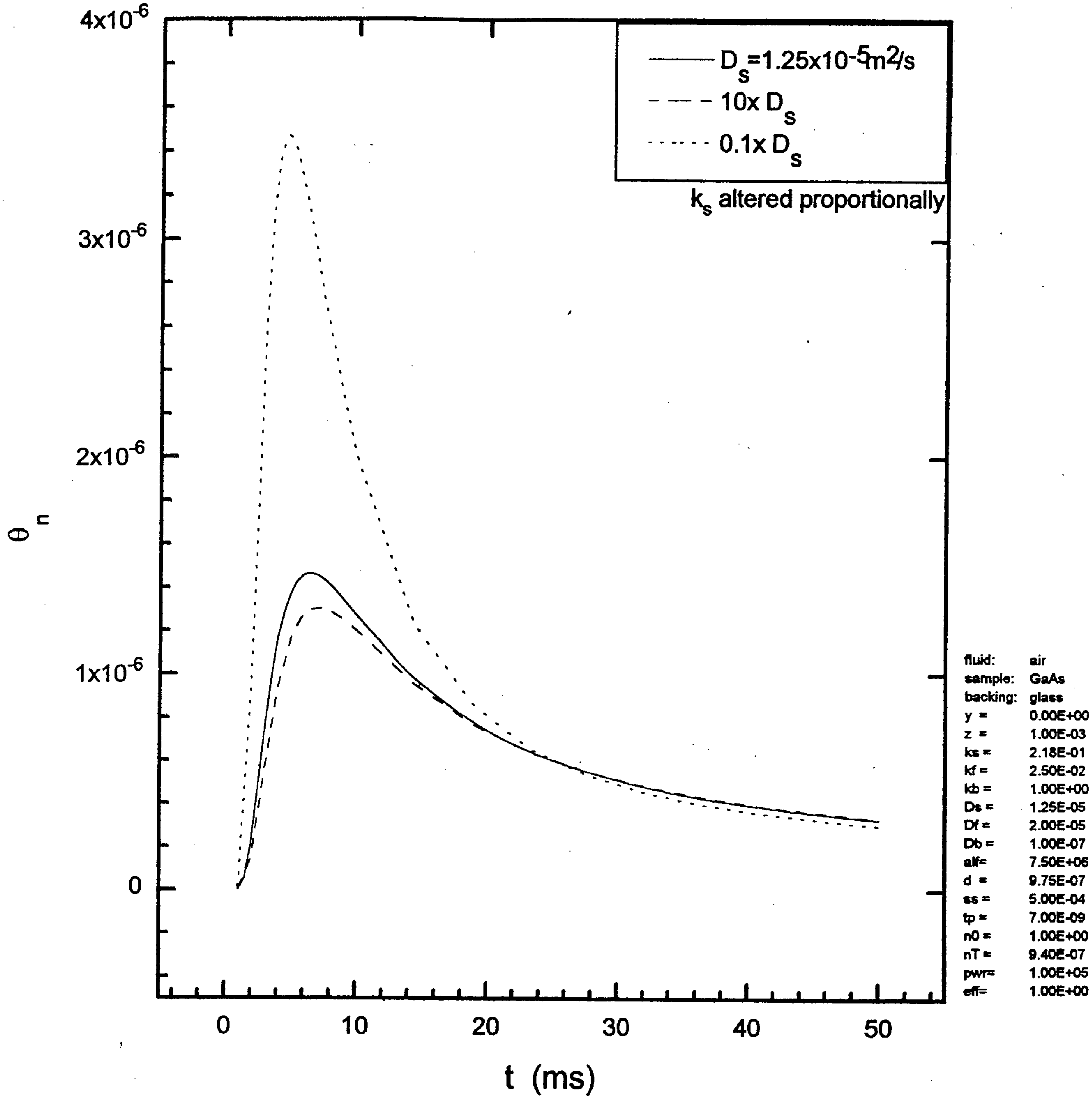

Fig. 2 This graph shows how the normal deflection signal is affected by a change in the sample diffusivity.

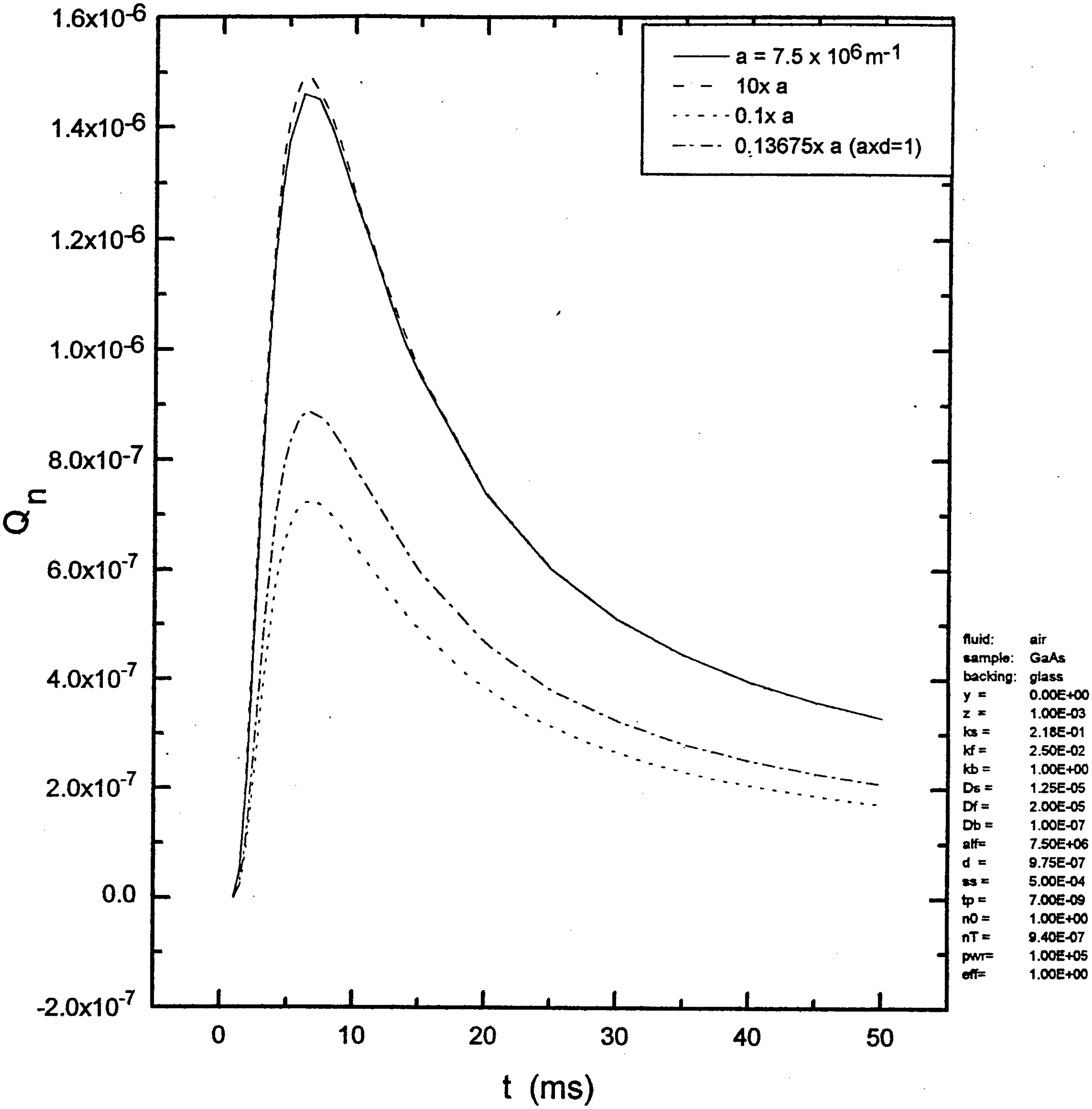

Fig. 3 The normal deflection signal for variations in the absorption coefficient.

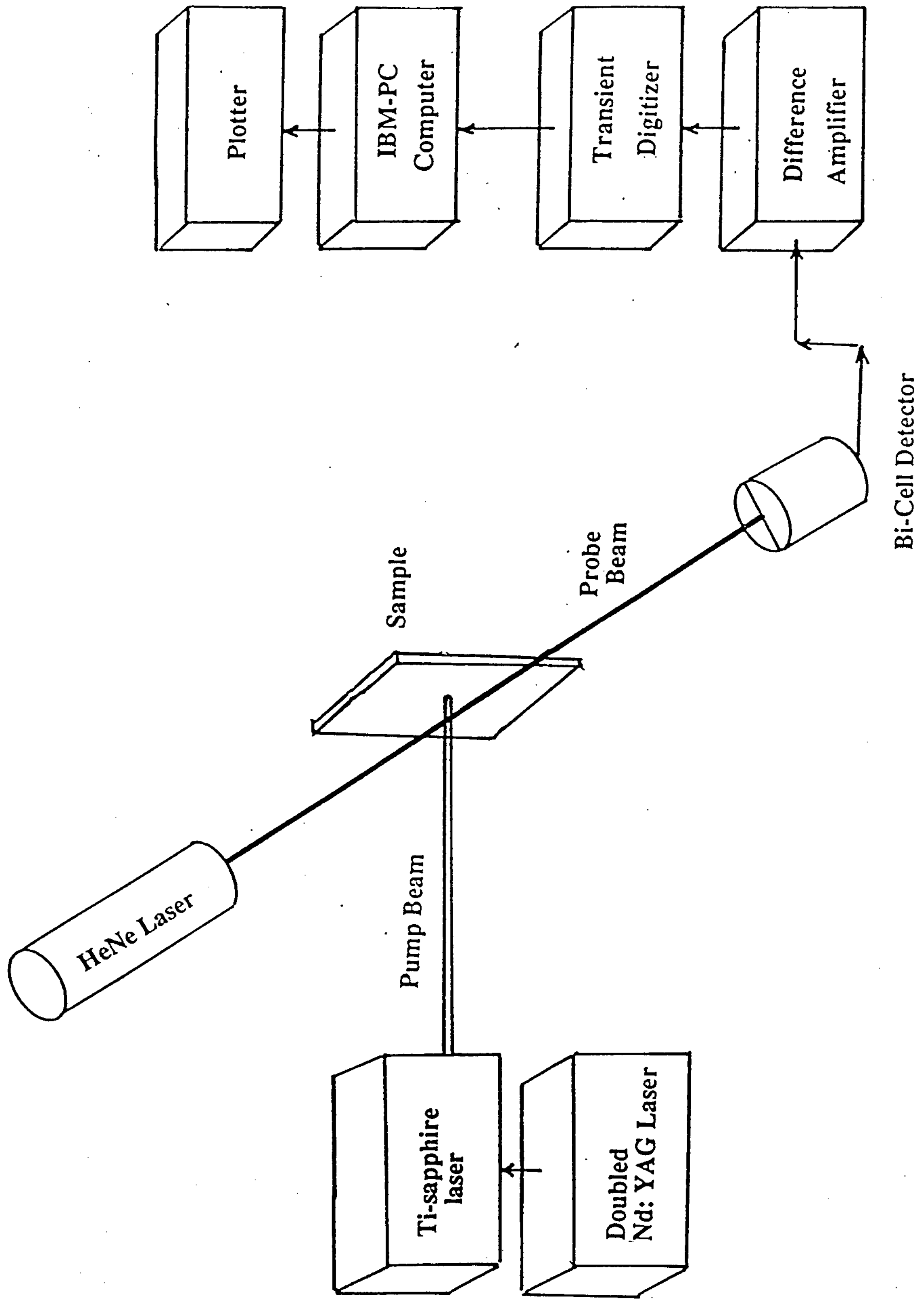

Fig. 4 Schematic representation of the experiment with pulsed laser excitation

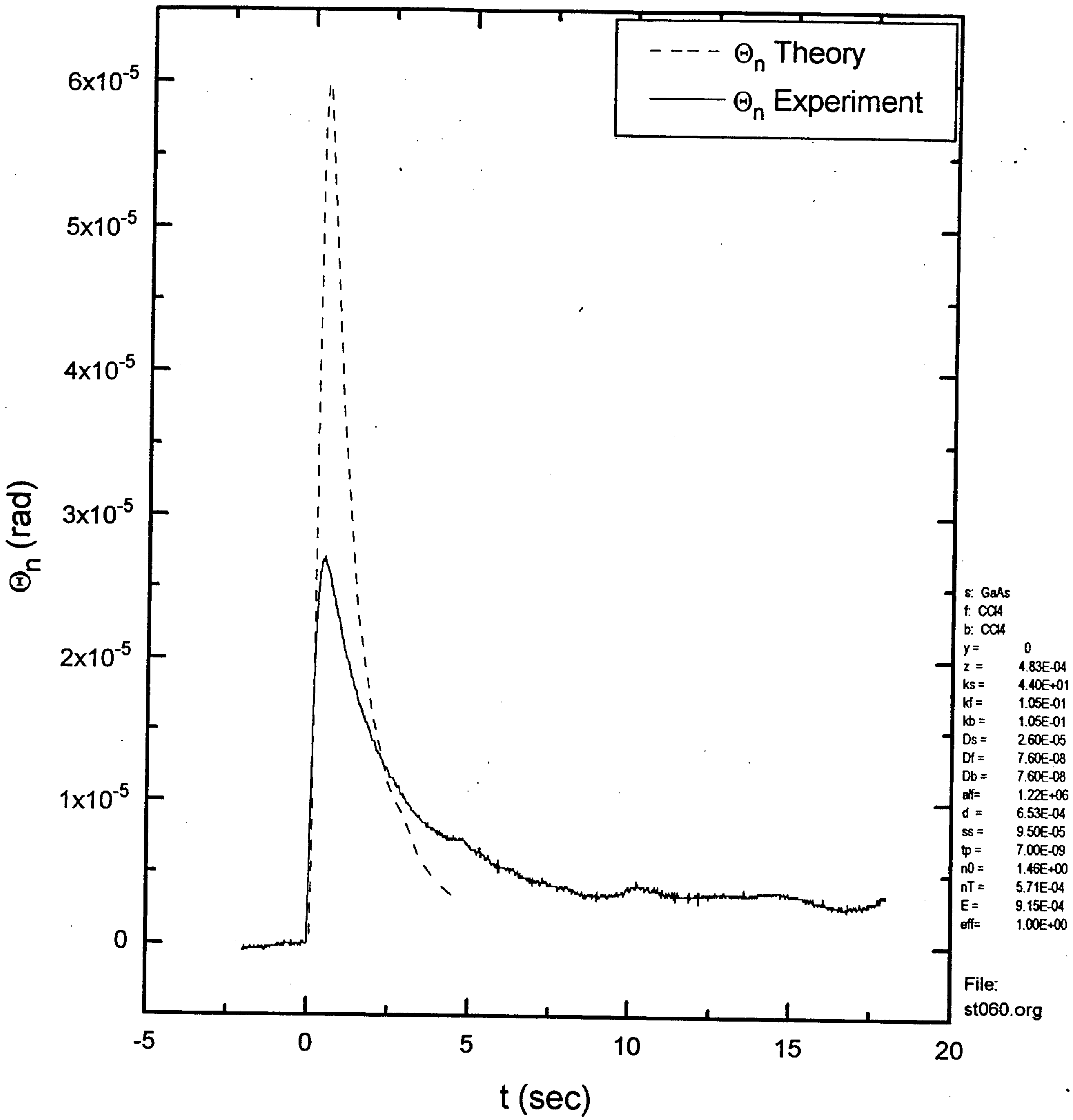

Fig. 5 Experimental (solid line) results and theoretical predictions (dashed line) for the normal component of the deflection signal in a wafer of GaAs.

DF (m^2/s)	= 7.56E-08	L (μm)	= 652.8	PWR (mW)	= 100
DS (m^2/s)	= var	F (Hz)	= 4	ALPHA (1/m)	= var
DB (m^2/s)	= 7.56E-08	A (μm)	= 1000	dn/dT (1/K)	= 3.90E-04
KF (W/mK)	= 1.03E-01	Y (μm)	= 0	L of I	= 10000
KS (W/mK)	= var	Z (μm)	= 431.8		
KB (W/mK)	= 1.03E-01				

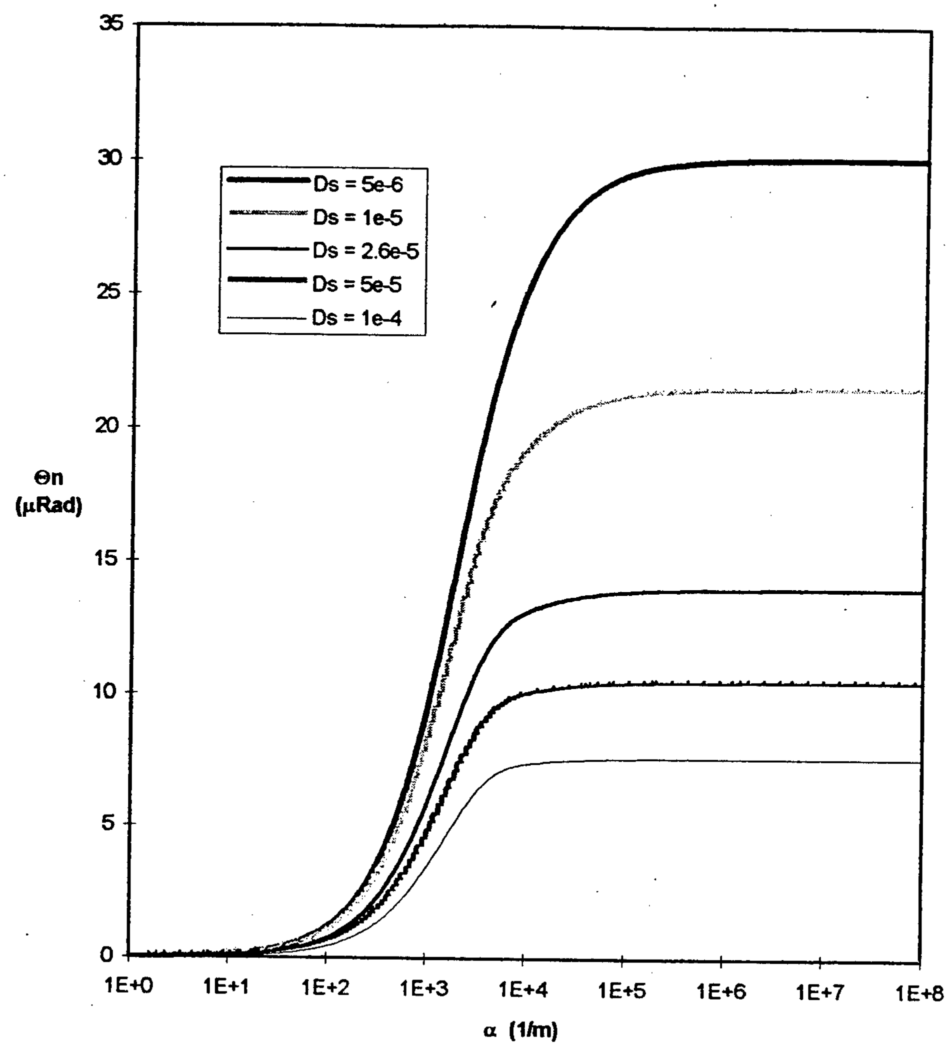

Fig. 6 Normal deflection versus the absorption coefficient with different values for the sample thermal diffusion constant and CCl_4 as the fluid.

DF (m^2/s) = 7.56E-08 L (μm) = 652.78 PWR (mW) = 100
DS (m^2/s) = var F (Hz) = 4 ALPHA (1/m) = var
DB (m^2/s) = 7.56E-08 A (μm) = 1000 dn/dT (1/K) = 3.90E-04
KF (W/mK) = 1.03E-01 Y (μm) = 0 L of I = 10000
KS (W/mK) = var Z (μm) = 431.8
KB (W/mK) = 1.03E-01

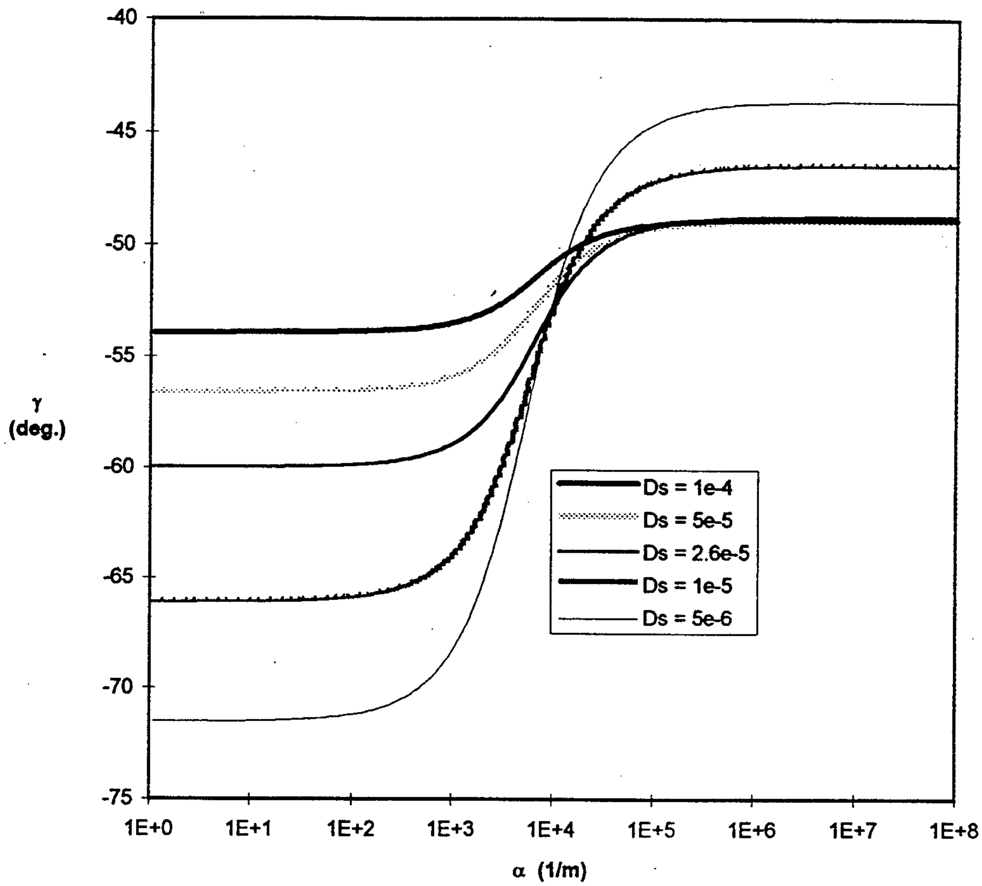

Fig. 7 Phase of the normal deflection versus the absorption coefficient for different values of the sample thermal diffusion constant and CCl_4 as the fluid.

DF (m^2/s) = 7.56E-08	L (μm) = 652.78	PWR (mW) = 100
DS (m^2/s) = 2.60E-05	F (Hz) = var	ALPHA (1/m) = 5.00E+06
DB (m^2/s) = 7.56E-08	A (μm) = 1100	dn/dT (1/K) =
KF (W/mK) = 1.03E-01	Y (μm) = var	L of I = 9090.91
KS (W/mK) = 44	Z (μm) = 431.8	
KB (W/mK) = 1.03E-01		

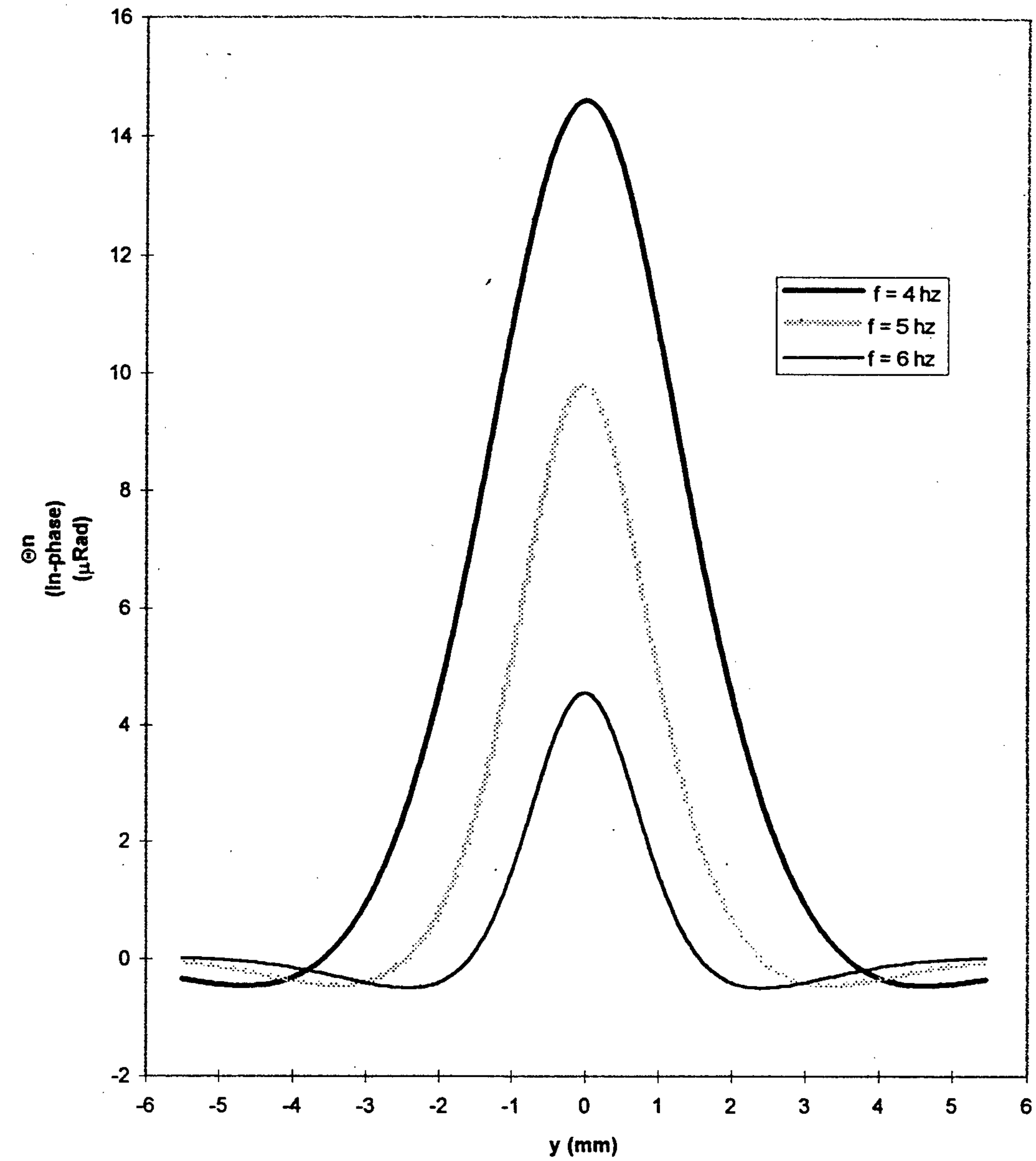

Fig. 8 The theoretical results for the in-phase portion of Θ_n versus y for 4, 5 and 6 Hz respectively.

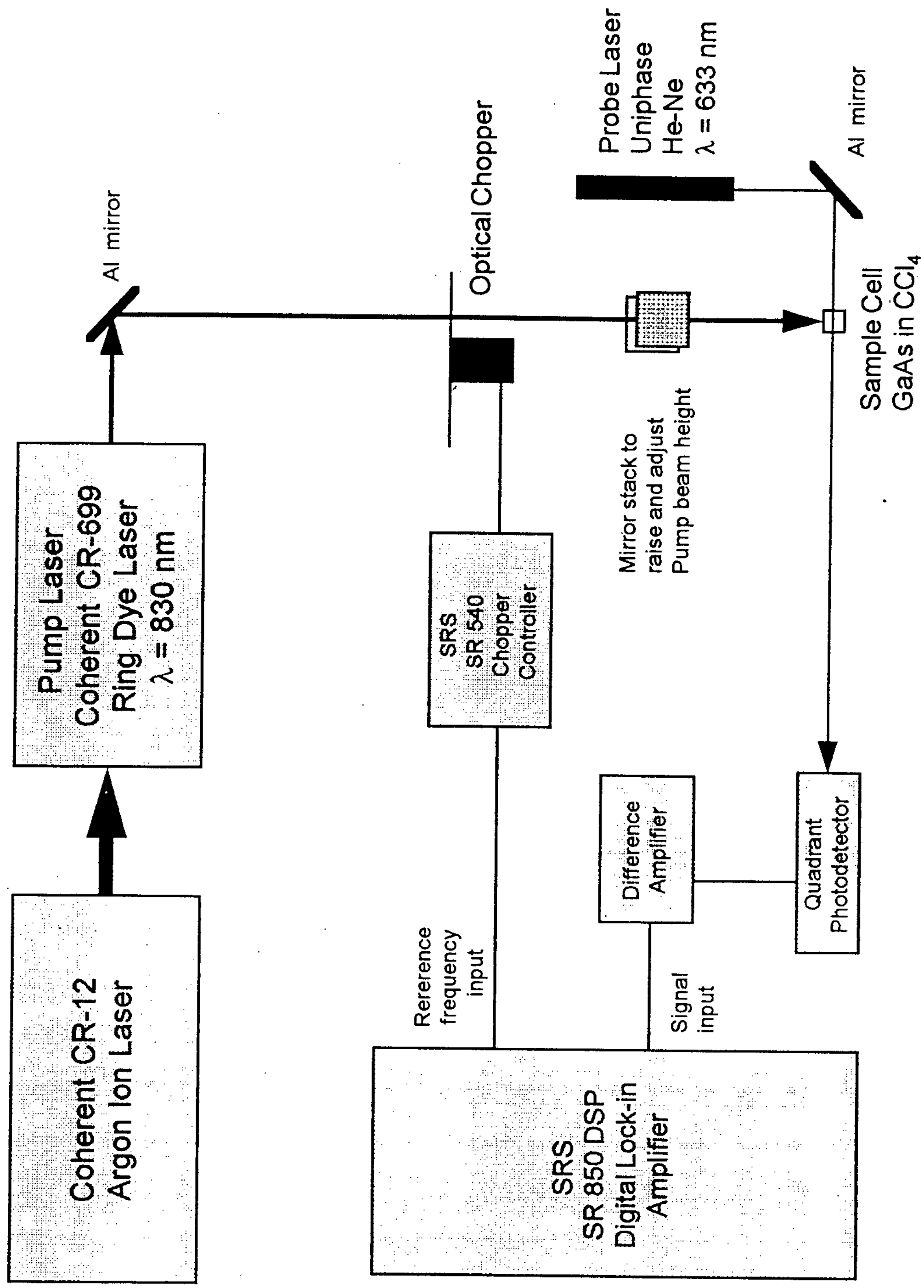

Fig. 9 Equipmental arrangement for a PTDS experiment with a CW pump laser and a solid sample.

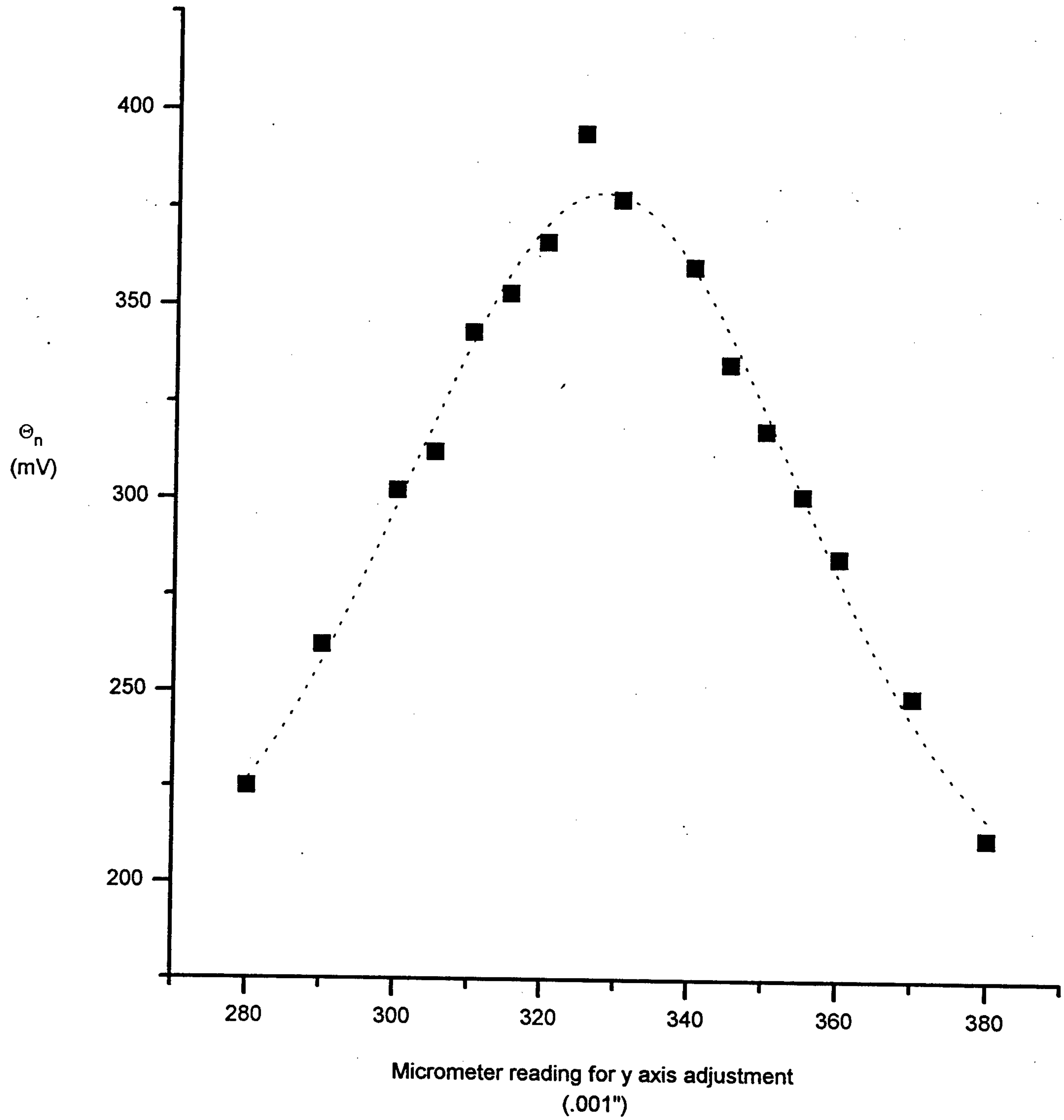

Fig. 10 Plot of the raw data for the normal deflection versus the y axis position for a GaAs sample in CCl_4.

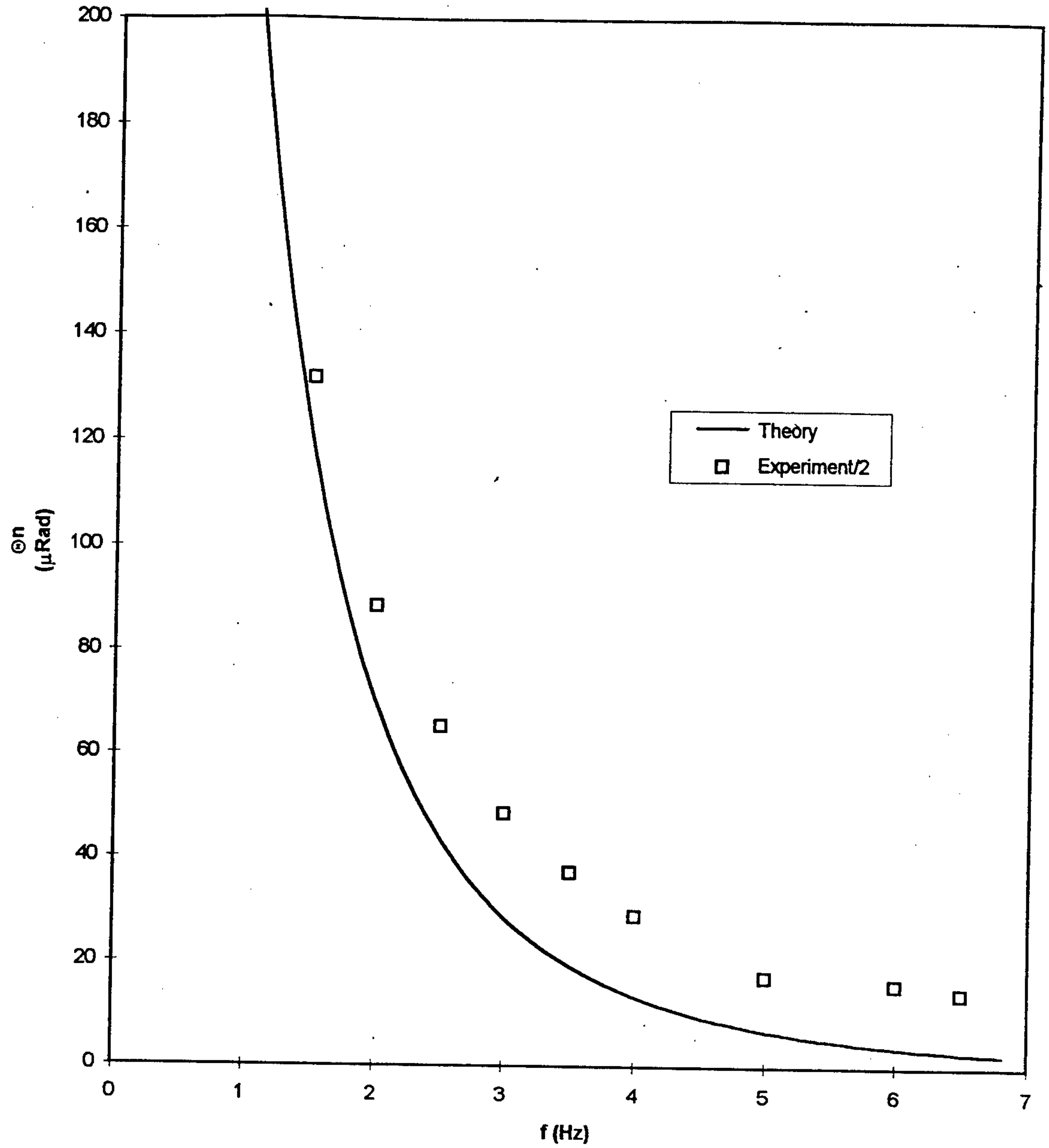

Fig. 11 Experimental and theoretical results for the normal deflection versus the modulation frequency. The experimental data is reduced by a factor of 2.

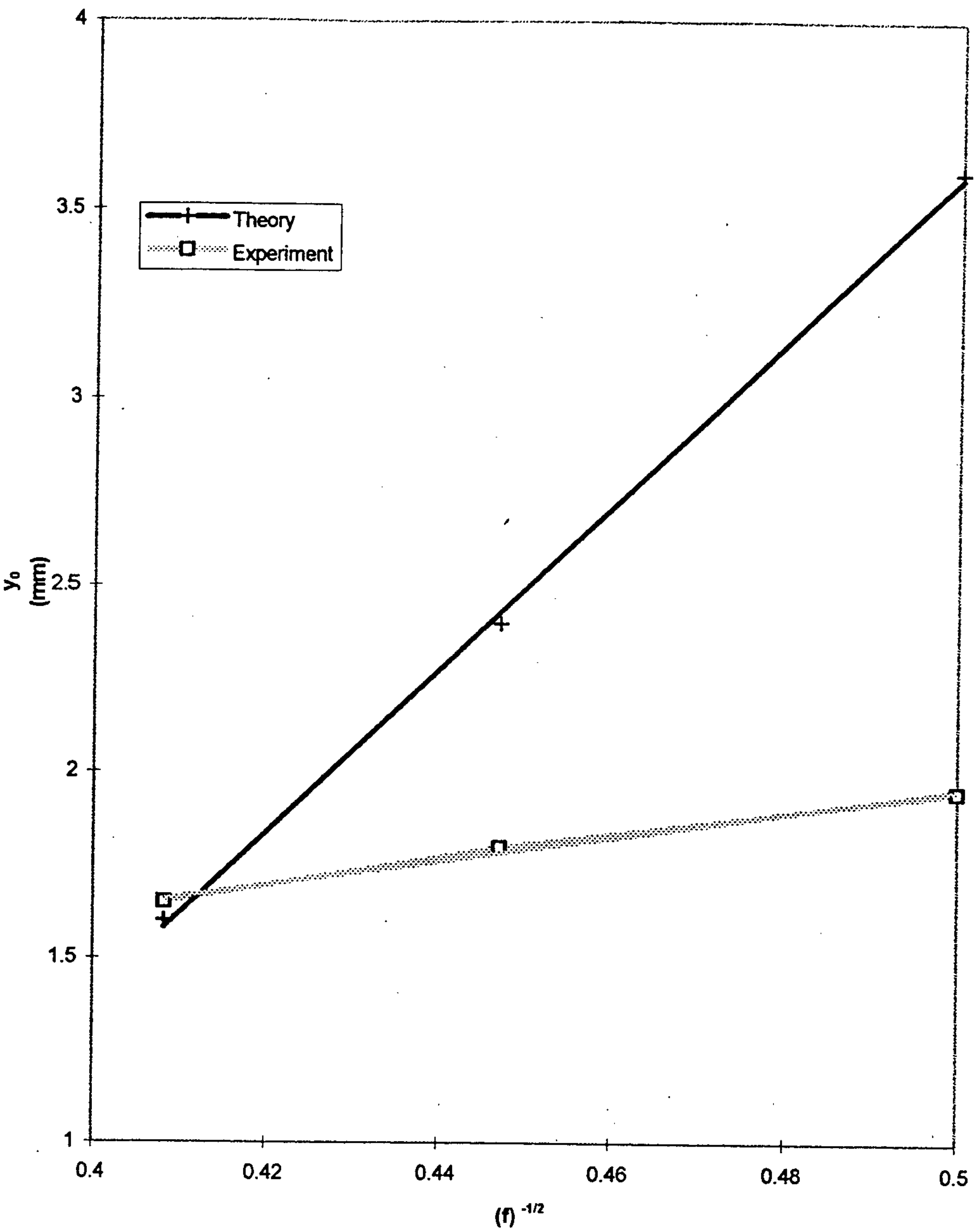

Fig. 12 The theoretical and experimental values for the crossing points of the in-phase portion of Θ_n versus $(f)^{-1/2}$ each shown with a linear fit line.